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TRANSLATION

OPTICAL DETERMINATION OF THE CONENTRATION OF WATER DROPS IN THE ATMOSPHERE DEPENDING ON THE RADIUS

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UNEDITED ROUGH DRAFT TRANSLATION

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English Pages: 17

Source: Aktinometriya i atmosfernaya optika, Leningrad, Gidrometeoizdat, 1961, pp. 218-227.

SC-1731
SOV/169-62-0-4-19/103

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OPTICAL DETERMINATION OF THE CONCENTRATION OF WATER DROPS IN THE ATMOSPHERE DEPENDING ON THE RADIUS

G. P. Gushchin

Starting from Mie's law and using the numerical values of the scattering function for water, ~~the author gives~~ a system of linear equations, determining for n wavelengths the extinction caused by a mixture of droplets with n different radii. After solving the system of equations, ~~we obtained~~ all n of the concentration corresponding to n radii.

In spite of the fact that the system of equations is given for droplets of pure water it was shown that it can be used with sufficient accuracy for droplets with a refractive index up to 1.4.

STUDY OF ATMOSPHERIC AEROSOLS

1. A systematic study of atmospheric aerosols by various methods and instruments is being carried out in the Departments of Aerological and Radiation Investigation at the A. I. Voyeykovo Main Geophysical Observatory (GGO). The present article describes the first part of an investigation which was performed by the ozonometric team of the GGO on the study of aerosols.

This investigation was divided into ground and aircraft observations. The task of the investigations included the following points:

a) To obtain systematic data on the spectrum optical thickness of aerosols in the ultraviolet and visible regions of the spectrum, to obtain the seasonal variation of the optical thickness of aerosols, and to study the relation between certain meteorological elements and the optical thickness of aerosols.

b) To obtain systematic information on the sizes and number of aerosol particles forming haze in the atmosphere under natural conditions during the annual cycle.

c) To obtain data on the vertical structure of the aerosol layer on a clear day by means of aircraft soundings of the atmosphere.

During 1957-1958 a total of about 5000 measurements of the optical thickness, size, and number of aerosol particles were made.

Three different instruments were used for the measurements:

a) A Dobson spectrophotometer whose optical portion was a double quartz monochromator with three constant slits.

b) A Scholtz condensation nucleus counter which was operated from an aircraft by scientific associate A. L. Dergach of the GGO.

c) An aircraft electrophotometer with light filters, fabricated at the GGO.

Heretofore in the overwhelming majority of cases the spectral properties of aerosols were studied by means of instruments with light filters or by means of spectrographs with singlefold resolution. The use in our case of a double quartz monochromator with high monochromatic qualities and a photoelectric system enabled us to increase frequently the accuracy of the measurements and to facilitate deciphering of the obtained data. For aerosol measurements we usedtwo permanently fixed slits of the monochromator which were installed in the instrument

especially to study aerosols; the first slit distinguished light of wavelength of 332 mμ and the second slit 456 mμ.

The spectrum intervals distinguishable by both slits were identica and equaled 3 mμ. Direct determination of the light intensity was found for a wavelength of 456 mμ. For the wavelength 332 mμ the light intensity was measured relative to the wavelength 456 mμ by means of a neutral optical wedge. The use of fixed slits and an optical wedge enabled us to obtain a high homogeneity and reproducibility of the obtained data.

In addition to the above indicated two slits, we used a third slit with a wavelength of 314 mμ which coupled with the 332 mμ slit served to measure atmospheric ozone.

The instrument provided for temperature compensation and the possibility of a rapid check of wavelengths by means of mercury-helium lamp. The sun was sighted by means of a quartz prism, wherein we used the method of an artificial fixed light source which was a dull quartz plate, scattering the light, installed in front of the entrance slit of the instrument.

2. The optical thickness of the aerosols was determined by measuring the intensity of direct sunlight in two narrow regions of the spectrum with centers of 332 and 456 mμ at different heights of the sun. The value of extraterrestrial solar radiation in selected regions of the spectrum was determined by the Bouguer-Langley method by constructing graphs of the logarithm of the intensity as a function of the air mass.

Over the course of two years we accumulated a large amount of statistical material on the distribution of extraterrestrial solar radiation in selected regions of the spectrum and determined their average values. This helps us to increase considerably the accuracy

of the measurements.

For the calculations we used Bouguer's formula

$$I_\lambda = I_{\lambda,0} 10^{-(\beta_\lambda + \delta_\lambda) m}, \tag{1}$$

where I_λ is the intensity of direct sunlight of wavelength λ at the level of the instrument; $I_{\lambda,0}$ is extraterrestrial intensity of sunlight; β_λ is the optical thickness of a pure atmosphere; δ_λ is the optical thickness of the aerosol layer; m is the air mass.

It follows from formula (1) that the optical thickness of the aerosols equals

$$\delta_\lambda = \frac{\lg I_{\lambda,0} - \lg I_\lambda}{m} - \beta_\lambda. \tag{2}$$

A correction for the ozone content in the atmosphere was introduced when using a wavelength of 332 mμ. However, this correction on the average did not exceed 10% of the optical thickness of the aerosols and was almost a constant value.

3. On the basis of the measurements of the optical thickness of aerosols in the indicated two regions of the spectrum, we determined the distribution of aerosol particles by radii in a verticle column of air with a cross-section of 1 cm^2.

For this purpose we made the following assumptions:

a) Atmospheric aerosols are spherical particles of different sizes with a refractive index of 1.33 (the refractive index of water);

b) The distribution of aerosol particles by radii satisfies a certain preselected distribution function depending on two parameters.

It was shown that under these conditions the relationship of the two indicated optical thicknesses of aerosols measured simultaneously is a function of one parameter which enters into the selected distribution function. From the value of the ratio of optical thicknesses

of the aerosols, we at first found the first parameter of the distribution function and then from the value of this parameter and the optical thickness we found the second parameter and thus determined the size distribution of the aerosol particles.

It follows from the Mie theory that the optical thickness (decimal) of aerosols for the two selected wavelengths λ_1 and λ_2 equals:

$$\delta_1 = 0{,}434\pi \sum_a a^2 \Delta N_a E(y_1), \tag{3}$$

$$\delta_2 = 0{,}434\pi \sum_a a^2 \Delta N_a E(y_2), \tag{4}$$

where $y_1 = \frac{2\pi a}{\lambda_1}$ and $y_2 = \frac{2\pi a}{\lambda_2}$. Here a is the variable radius of particles in centimeters; ΔN_a is the number of particles in a vertical column of air with a cross section of 1 cm^2, the radius of which lies within the limits from a to a + Δa; E(y) is the function of Houghton and Chalker.

As one of the distribution functions we used the relationship obtained by Foitzik for fog,

$$\Delta N_a = N_r e^{-c\left(\log\frac{a}{r}\right)^2} \Delta a, \tag{5}$$

where r and N_r are parameters; c is some constant, $\Delta a = 0.05\ \mu$ [1].

It follows from (3), (4) and (5) that

$$\frac{\delta_1}{\delta_2} = \frac{\sum_a a^2 e^{-c\left(\log\frac{a}{r}\right)^2} E(y_1)}{\sum_a a^2 e^{-c\left(\log\frac{a}{r}\right)^2} E(y_2)}, \tag{6}$$

i.e.

$$\frac{\delta_1}{\delta_2} = f(r). \tag{7}$$

This last relationship (7) shows that the ratio of the optical thicknesses for the given λ_1 and λ_2 depends only on the parameter r.

The function f(r) for c = 27 and c = 50 was calculated and was shown in Fig. 1. We should note that the two curves in Fig. 1 for c = 27 and c = 50 are comparatively close to each other.

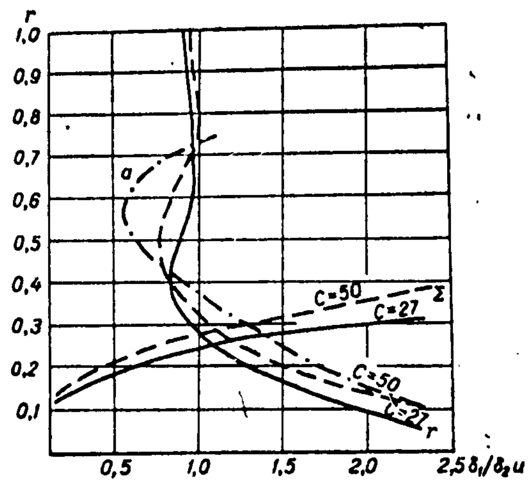

Fig. 1. Graph for determining the parameters r and N_r of the size distribution function of aerosol particles $\Delta N_a = N_r e^{-c\left(\log \frac{a}{r}\right)^2} \Delta a$ as a function of the ratio of the optical thicknesses of aerosols $\frac{\delta_1}{\delta_2}$ for $\lambda_1 = 332$ mμ and $\lambda_2 = 456$ mμ. The value of N_r is found from the expression $N_r = 7.34 \cdot 10^7 \frac{\delta_1}{\Sigma}$. Curve a served to determine the radius of particles of monodispersion sols with a refractive index of 1.33.

An analogous function was calculated for particles of identical sizes and is shown for comparison in Fig. 1 (curve a). By means of curve a (Fig. 1) we determined the particle sizes of monodispersion aerosols. This curve is also close to the two curves calculated for

c = 27 and c = 50.

The second parameter N_r of function (5) on the basis of (3) was found from the following expression:

$$N_r = \frac{\delta_1}{0{,}434\pi \sum_a a^2 e^{-c\left(\log\frac{a}{r}\right)^2} E(y_1)\,\Delta a}. \tag{8}$$

To facilitate calculations when finding N_r we calculated the auxiliary function

$$\Sigma(r) = \frac{1}{\sum_a a^2 e^{-c\left(\log\frac{a}{r}\right)^2} E(y_1)}, \tag{9}$$

the graph of which for c = 27 and c = 50 is shown in Fig. 1.

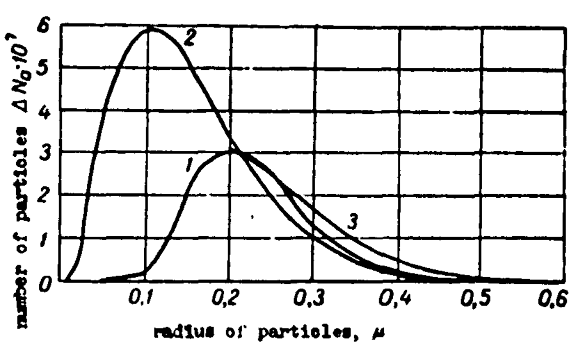

Fig. 2. Distribution of the number of aerosol particles by sizes with an interval Δa = 0.05 μ in a vertical column of air with a cross section of 1 cm^2. Voyeykovo village, June 26, 1958, 0940. 1) calculated by formula $\Delta N_a = N_r e^{-27\left(\log\frac{a}{r}\right)^2}\Delta a$; 2) calculated by formula $\Delta N_a = Aa^2 \cdot 10^{-ka}\Delta a$. 3) obtained from the data of G. Dessens on September 28, 1946 (relative units).

For comparison we used another distribution function

$$\Delta N_a = Aa^2 \cdot 10^{-ka}\Delta a, \tag{10}$$

where k and A are parameters.

In comparison with (5) it has the advantage that it is free from

the incompletely determined parameter c.

On the basis of these measurements on the optical thickness of aerosols, we determined the parameters k and A like in the preceding case by means of function (10) and found the distributions of particles by radii with the same interval $\Delta A = 0.05\ \mu$. Both types of distributions obtained by formulas (5) and (10) which pertain to one and the same instant of time differ from each other when $a < 0.2\ \mu$, but when $a > 0.2\ \mu$ they virtually coincide (Fig. 2).

4. From the obtained particle distribution we calculated the spectrum optical thickness of the aerosols (decimal) in the range of the wavelengths from 200 to 800 mμ. For this purpose we used formula (3) wherein parameter y was varied due to the change in radius a and because of the change in wavelength λ.

The values of the optical thickness of aerosols in various regions of the spectrum prove to be close for two distribution of particles by radii calculated by different formulas (5) and (10), but measured at one and the same time (Fig. 3). Hence we can conclude that the spectrum optical thickness of aerosols is practically independent of the distribution of small aerosol particles with $r < 0.2\ \mu$.

5. By means of formula (2) and curve r for c = 27 (Fig. 1) we estimated the errors of the values of δ and r obtained as the result of the measurements with the Dobson spectrophotometer. The relative error of this value was 3-5% for average values of the optical thickness of aerosols δ.

The relative error of the parameter r depend on values of δ and r. The average relative error of the value of r was 10-15%.

The relative error of the values of N_r, as the calculations show, was approximately twice that of the value of r.

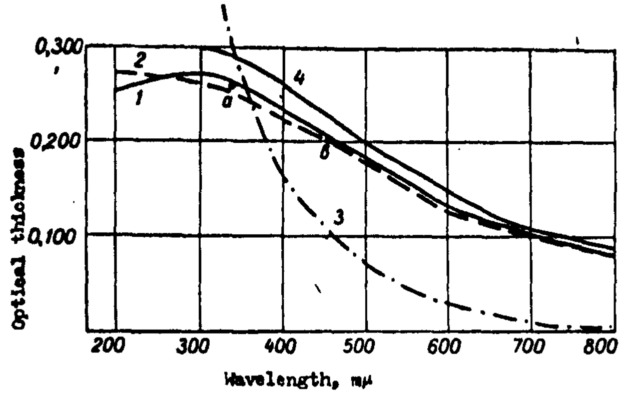

Fig. 3. The optical thickness of aerosols (1), (2) and the optical thickness of pure atmosphere (3) as a function of wavelength. Voyeykovo village, June 26, 1958, 0940. Curve 1 was derived for the distribution $\Delta N_a = N_r e^{-n\left(\log\frac{a}{r}\right)^2}\Delta a,$ curve 2 for the distribution $N_a = Aa^2 \cdot 10^{-ka}\Delta a.$ Points a and b were obtained as a result of direct measurement of the optical thickness of aerosols and are the reference points when calculating curves 1 and 2. Curve 4 was obtained from the data of O. I. Popov for a horizontal layer of aerosols (relative units).

6. Aircraft observations included measurement of the index of aerosol attenuation γ in km^{-1} and the optical thickness of aerosols δ_h at various heights in the atmosphere.

The aircraft measurement of these magnitudes was done by the electrophotometer with light filters designed at the GGO, which enabled us to measure the intensity of direct solar light with a wavelength of 370 mμ.

The instrument consisted of the following main parts: light filter (combination of UFC-2 + SZS-18), magnesium photocell, d-c amplifier on a 1NZS tube, control panel.

For the reading we used the M-24 micro-ammeter, a sight was used for shooting the sun.

The instrument was equipped with a monitor device of the photoelectric system; this was a specially built-in 6.3 v bulb. The linearity of the instrument was checked by means of neutral light filters. The linearity proved to be sufficiently good for the entire scale of the instrument.

7. Calculation of the attenuation index of the atmosphere at height h km was done by formula

$$\sigma_h = \frac{\log \frac{I_{h_1}}{I_{h_2}}}{m_c(h_1 - h_2)} \text{ км}^{-1}, \tag{11}$$

where I_{h_1} is the intensity of direct sunlight at height h_1 km and I_{h_2} is the same at height h_2 wherein we took $h_1 - h_2 = 1$ km and $h = h_1 - {}^1/_2$ km.

Formula (11) is easily derived from the Bouguer formula if the atmosphere is divided into layers 1 km thick.

It follows from formula (11) that the maximum relative error σ equals

$$\frac{\Delta\sigma}{\sigma} = \frac{\Delta m}{m} + \frac{\Delta(h_1 - h)}{h_1 - h} + \frac{0.434}{\log\frac{I_h}{I_{h_1}}}\left(\frac{\Delta I_h}{I_h} + \frac{\Delta I_{h_1}}{I_{h_1}}\right). \tag{12}$$

Calculation of the relative error by formula (12) for average conditions yielded a value of about 25% for $\frac{\Delta\delta}{\delta}$. The aerosol attenuation index γ_h km -1 was determined by subtracting from σ_h the attenuation index of a layer of pure air 1 km thick.

In addition to the aerosol attenuation index, we calculated the optical thickness of aerosols at different heights in the atmosphere δ_h. Formula (2) was used for calculating δ_h.

8. The following conclusions can be made from the results of measuring the values δ, r, and $N_r \Delta a$ at Voyeykovo in 1958:

a) The magnitude of the optical thickness of aerosols δ (decimal)

for $\lambda = 332$ mμ varied during the year from a value close to zero to 0.5; the value δ' for $\lambda = 456$ mμ, from that close to zero to 0.4 wherein $\delta > \delta'$ was systematic. The values of δ and δ' increased from winter to summer. For example, in February the mean value of δ was 0.078 and δ' was 0.050, whereas in June it was respectively 0.173 and 0.115.

A seasonal change in the optical thickness of the aerosols with a summer maximum and winter minimum exists at Voyeykovo.

b) The diurnal variation (during the daytime) of the optical thickness of the aerosols was obtained from the measurements at Voyeykovo on clear days. The maximum of the optical thickness was usually observed during the noon hours and the minimum during the morning and evening hours. As an example we can cite the observation of June 30, 1958. Between 0600 and 0700 hours Moscow time the average value of the optical thickness of aerosols for 456 mμ was 0.07, between 1300 and 1400 hours 0.20, and between 1800 and 1900 hours 0.08.

c) From numerous determinations (1070 cases) of the optical thicknesses of aerosols δ and δ' when δ and δ' were greater than 0.050, we found the average value of the coefficient α in the Angstrom formula $\delta = \text{const}\ \lambda^{-\alpha}$. This value equals $\alpha = 1.3$.

d) The value of r (the radius of particles at the distribution maximum) from formula (5) was a very stable magnitude during the year and equaled on the average 0.17 μ. In February the average value of r was 0.15 μ and in June 0.16 μ. The same value r, but found from formula (10), was slightly less and averaged 0.08-0.1 μ (Fig. 2). This fact supports the assertion of Gilbert that the majority of atmospheric particles with sizes from 0.05 to 1 μ consists of supercooled water and that a refractive index of 1.33 is applicable to them [6].

e) The values of $N_r \Delta a$ (number of particles at the distribution maximum) when $\Delta a = 0.05$ μ varied from 10^7 to 10^8 particles. In

February the average value of $N_r\Delta a$ equaled $3.5 \cdot 10^7$ and in June $7 \cdot 10^7$.

f) The value of r generally increased with an increase of δ and δ'.

9. As a result of the measurements at Voyeykovo and subsequent calculations by formulas (5) and (10), we obtained a series of curves of the size distribution of aerosol particles. An examination of these curves leads to the conclusion that the measured particles had sizes (radii) from 0.05 to 0.7 μ.

A comparison of our distribution curve with curves obtained by Dessens [5] by other methods indicates a similarity between these curves when $a > 0.2$ μ (Fig. 2).

The obtained results on the measurement of the sizes of aerosol particles are interesting in that it was possible to measure particles whose sizes are inaccessible for microscopic measurement without interference in the medium.

In addition the proposed method permits us to measure particle sizes in a verticle column passing through the entire atmosphere, which is very important when calculating the passage of radiation through the atmosphere.

By means of the distribution curves obtained at Voyeykovo, we calculated by formula (3) the values of the optical thickness of aerosols δ_λ as a function of wavelength. On the basis of these curves and on the basis of measuring the total amount of ozone in the atmosphere, we plotted appraisable curves of the energy distribution in the solar spectrum at the earth's surface in the region 300-500 mμ. For this purpose we used the data of Jonson on the extraterrestrial solar spectrum [7]. It required only about 1 min to obtain initial experimental data when plotting such a curve of the energy distribution in the solar spectrum at the earth's surface.

10. It was established upon measuring the optical thickness of aerosols from the earth's surface that an intimate relation does not exist between the inclined transparency of the atmosphere (in the direction toward the sun) and the horizontal visual range. A number of cases were noted (May 23, June 26, June 30, 1958, etc.) where the optical thickness of the aerosols toward the sun increased with an increase in the horizontal visual range in the southern sector of Voyeykovo.

11. It was established upon measurements from the earth and from an aircraft that the appearance or formation of cloudiness outside the solid angle of the instrument in a number of cases led to a noticeable increase of the optical thickness of the aerosols. The disappearance of cloudiness led to the opposite result. This fact permits us to speak about the existence of invisible clouds or invisible continuations of ordinary clouds consisting of particles with a radius of 0.05-0.7 μ.

12. As a result of the aircraft measurements of the aerosol attenuation index γ, optical thickness of aerosols δ, and the concentration of condensation nuclei N at various altitudes, the following conclusions were derived:

a) In each individual case the magnitude γ characterizing the vertical distribution of aerosols is not governed by the exponential law upon a change of h (Fig. 4). Aerosol layers are quite frequently observed at heights of 2-6 km. Similar results in certain individual cases were obtained at the TsAO by G. T. Faraponova (Fig. 4) in 1957 [4] and by A. G. Laktionov in 1958 [2] wherein the measurements of the latter were carried out for particles whose radius was greater than 4 μ. This circumstance confirms the postulation that on clear days the atmosphere has a nonhomogenous structure and that invisible clouds

consisting of particles with a radius of 0.05-0.7 μ are in the atmosphere.

b) At a height of 6 km the average value of the optical thickness of aerosols δ_h (decimal) for λ = 370 mμ during measurement (10 flights) in a clear sky over various points of the USSR was stable and equaled 0.03.

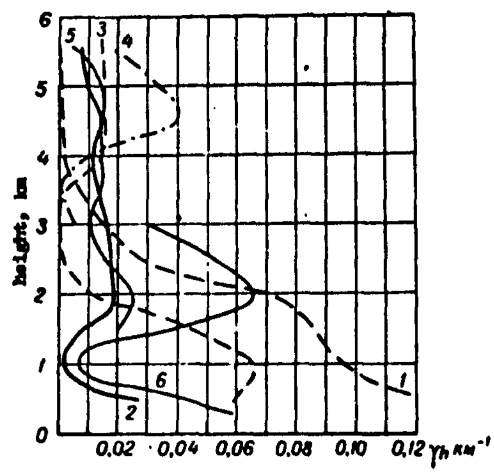

Fig. 4. Measurement of the aerosol attenuation index (decimal) γ with height. 1) Leningrad region April 9, 1958; 2) Leningrad region, June 19, 1958; 3) Leningrad region, July 30, 1958; 4) Mineral'nyye Vody, October 7, 1958; 5) Aktyubinsk, October 28, 1958; 6) Data of Faraponova, TsAO, June 20, 1956.

It was found from the measurement at Voyeykovo and interpolation that the mean annual value of the optical thickness of aerosols for λ = 370 mμ measured from the earth's surface is 0.125.

Hence it follows that above 6 km a still considerable portion of the aerosols, comprising on the average one-fourth of the entire aerosol layer in the case of measurements with an optical instrument (i.e., for particles with a radius of 0.05-0.7 μ), is located in the atmosphere. Values of δ_h greater than 0.03 at a height of 6 km were

noted on cloudy days (upper limit 3-5 km).

c) By measuring the concentration of aerosols by two different methods it is possible to establish that on clear days the number of fine particles in the lower troposphere measured by the Scholtz counter ($0.001 < r < 0.1\ \mu$) is greater than the number of large particles measured by an electrophotometer with light filters ($0.05 < r < 0.7\ \mu$) wherein the number of large particles with respect to all particles was on the average 1-5%. This result is in accord with the data of Junge on the distribution of aerosol particles by sizes in the lower troposphere [8].

d) A joint examination of the curves of the vertical distribution of condensation nuclei and the aerosol attenuation index leads to the conclusion that there is a correlation between these two curves (Fig. 5). However, the concentration of condensation nuclei decreases with height on the average more rapidly than the aerosol attenuation index.

To explain this somewhat unexpected fact we put forward the hypothesis that the large particles detectable by the electrophotometer in contrast to the fine particles detectable by the Scholtz counter for the most part have a condensation nature and a water shell. In those layers of the atmosphere where the relative humidity is high, the aerosol particles increased their size by condensation and are detected by the optical instrument. Layers of such particles on a clear day do not change into clouds due to the lack of moisture but are detected as layers of high haze. Upon an increase in humidity at the corresponding atmospheric levels these particles can form clouds. In view of the fact that the temperature in the troposphere drops with height and water vapor is propogated mainly from the bottom upward, suitable conditions for the formation of layers of large particles are

frequently present in the troposphere during the summer.

In conclusion I wish to thank R. G. Romanova and V. B. Laeksandrovich, co-workers of the A. I. Voyeykovo Main Geophysical Observatory for their help in fulfilling this work.

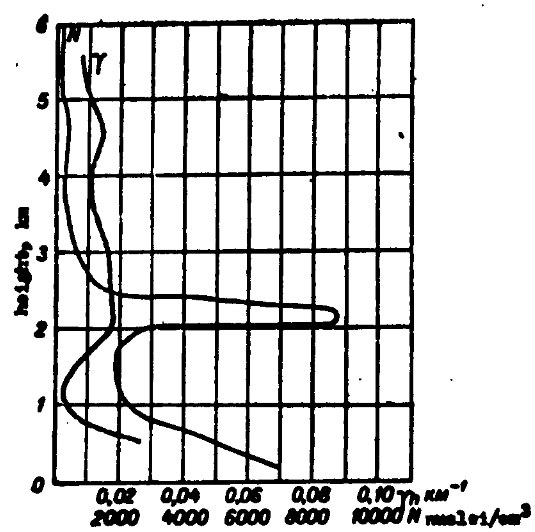

Fig. 5. Aerosol attenuation index γ in km^{-1} and the concentration of condensation nuclei N in $nuclei/cm^3$ at various atmospheric heights. Voyeykovo village, June 19, 1958.

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